

Fiber-optic-coupled, laser heated thermoluminescence dosimeter for remote radiation sensing

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We describe an optically transparent, laser heated, thermoluminescent glass dosimeter that is used for fiber-optic-coupled remote radiation monitoring. The glass consists of ZnS nanocrystallites embedded in a Vycor glass host, doped with Cu^{1+} and Nd^{3+} ions. An 807 nm, 1 W GaAlAs laser is used to rapidly heat microscopic regions, surrounding the Nd^{3+} ions, in the composite glass to stimulate blue-green thermoluminescence from metastable traps populated by exposure to ultraviolet or ^{60}Co γ radiation. [S0003-6951(96)03424-9]

Measurements of radiation levels in hazardous areas, such as nuclear waste sites or in difficult-to-access areas such as sampling wells for *in situ* ground water monitoring, require the development of sensitive remote radiation measurement techniques. Fiber-optic-based methods should be particularly amenable to these applications for several reasons, including: small size, mechanical flexibility, and optical signal transmission (as opposed to electrical). Several fiber-optic-based radiation sensing systems have been described that utilize radiation induced changes in the optical characteristics of the fiber such as reduced transmission as a result of darkening of the glass,¹ optical phase shifts due to heating,² or changes in the birefringence of a polarization-maintaining fiber.³ Another approach utilizes a traditional thermoluminescence (TL) radiation dosimeter phosphor, attached to the end of a multimode fiber-optic cable. The phosphor is heated by thermal diffusion from an adjacent laser heated absorber.⁴ Each of these fiber-optic-based techniques suffer from serious shortcomings that limit their practical application. The measurement of radiation induced darkening is a straightforward approach but is limited in both sensitivity and dynamic range. In addition, photodarkening is usually the result of photochemical damage and the process is not generally reversible. Phase shift measurements require the use of an interferometer with phase sensitive detection and feedback control, long fiber lengths, and complex signal processing techniques to achieve good sensitivity. The fiber-optic, laser-heated TL approach requires the use of a very thin layer of phosphor material because of problems associated with light scattering and inefficient heating by thermal diffusion. This limitation on the thickness of the dosimeter material ultimately limits the sensitivity of the method.

In this letter, we describe a remote, fiber-optic-coupled, laser heated TL glass radiation dosimeter that has advantages over previously reported fiber-based dosimeters. We recently reported the development of an optically transparent thermoluminescent glass material containing ZnS nanocrystals and Cu^{1+} ions. This material exhibited outstanding dosimetry characteristics, including a low fade rate, a linear dose

response of over six orders of magnitude, an energy response that extends from the ultraviolet ($\lambda < 300$ nm) to γ -ray energies, and a sensitivity similar to commercial TLD 100.⁵ The glow curve of the TL glass is particularly simple, consisting of two broad peaks located at 160 and 220 °C. We have also reported⁶ a modified TL glass composition that incorporated Nd^{3+} ions to absorb light from a semiconductor laser and utilized the absorbed light energy to internally heat the glass. Internal laser heating of the glass eliminated thermal diffusion problems associated with external absorbers and the optical transparency overcame the scattering limitations of traditional TL phosphors. As a result, the radiation sensitivity of the TL glass dosimeter can be enhanced simply by increasing the length or the diameter of the fiber.

The laser heated TL glass used in this work is a composite material that contains nanocrystalline ZnS particles and Cu^{1+} ions in a SiO_2 (Vycor) matrix, identical to that previously reported.⁵ Approximately 0.1%, by weight, of Nd^{3+} ion was added to the glass to accomplish internal laser heating. The Nd^{3+} ion is well suited for this application for several reasons: (1) it absorbs strongly at 807 nm (GaAlAs laser wavelength); (2) it deposits a significant amount of the absorbed light energy as heat in the glass matrix to stimulate TL; and (3) the absorption bands of the Nd^{3+} ion do not significantly overlap the thermoluminescence emission band and therefore do not substantially attenuate the signal. Glass rods, 6 mm in diameter, were hand-drawn into ~ 200 μm diam fibers using a hydrogen torch. An ~ 3 cm length of the TL fiber was joined to a commercial, 200 μm core diameter, multimode optical fiber using a fusion splicer.

A schematic of the fiber-optic-coupled, laser-heated thermoluminescence dosimeter is shown in Fig. 1. The 807 nm output of a 1 W diode laser array, with a 200 μm emitting aperture, was collimated using a microscope objective. The laser output was filtered with a Schott RG780 color glass filter to remove short wavelength light due to spontaneous emission. The laser light was directed through a dichroic mirror, selected to pass 807 nm light and reflect blue-green TL signal light, and imaged onto the input face of a 200 μm core diameter, 50 m long multimode optical fiber using a second microscope objective. The laser light was used to stimulate thermoluminescence from the ZnS:Cu,Nd

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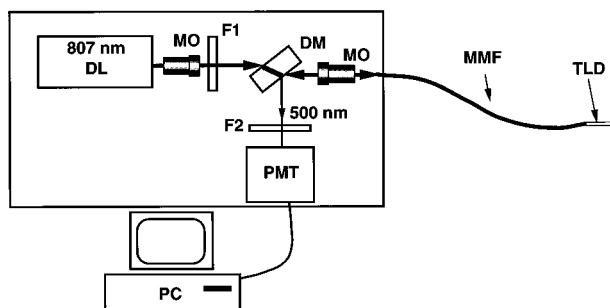


FIG. 1. Schematic diagram of the laser-heated fiber-optic-coupled thermoluminescence dosimeter. DL: diode laser; DM: dichroic mirror; MO: microscope objective; MMF: 200 μm core diam multimode optical fiber; TLD: thermoluminescent glass dosimeter fiber; F1, Schott RG780 color glass filter, F2: HOYA CM500 color glass filter; PMT: cooled photomultiplier tube.

glass fiber sensor that had been previously exposed to ionizing radiation. A fraction of the 500 nm, laser stimulated TL emission was trapped by total internal reflection and was directed back through the multimode fiber, collimated by the microscope objective and reflected by the dichroic mirror into a photomultiplier tube (PMT). A HOYA CM500 color glass filter was used to selectively block scattered diode laser light while passing the TL signal. The PMT was cooled and the signal was detected using photon counting and digitized. The fiber-coupled sensor was placed above a ^{60}Co γ -ray source providing an exposure rate of 5 R/min. The dose received was controlled by irradiating the sensor for a specific time interval. The absorbed dose was measured by switching the diode laser on to a preset cw power of 250 mW for a period of 10–20 s. The traps released by laser heating were annealed, thereby resetting the sensor for repeat use. A key feature of the remote dosimeter is the ability to anneal the glass optically, so that repeated, *in situ* measurements can be performed. The remote sensor described in this work has demonstrated the ability to accumulate absorbed dose over long periods of time and yield an accurate measure of the total absorbed dose upon laser readout. For many practical applications, however, such as patient dose verification during medical radiotherapy, real-time, *in situ* readout of the absorbed dose under conditions of continuous irradiation is desirable. The data presented in this work were obtained under conditions of continuous irradiation, therefore, the laser-off state corresponds with dose accumulation, while the laser-on state corresponds with dose readout and sensor annealing.

A laser heated TL signal obtained from a fiber dosimeter previously exposed to a ^{60}Co γ -ray dose of 4.56 Gy is presented in Fig. 2. In the figure the arrow indicates the time that the laser was switched on. This corresponded with an immediate (<0.01 s) increase in the signal level, followed by a decay of several seconds. This behavior is in marked contrast to previous studies of laser heated dosimetry⁴ in which the recorded signal is similar in structure to the glow curves observed using traditional, linear temperature ramp, contact heating methods. Prior laser heating methods involve raising the bulk temperature of the dosimeter at heating rates of over 1000 $^{\circ}\text{C}/\text{s}$. The laser heating observed in the present work is also quite rapid, however, measurement of the rise of the signal using a fast oscilloscope reveals that it follows exactly

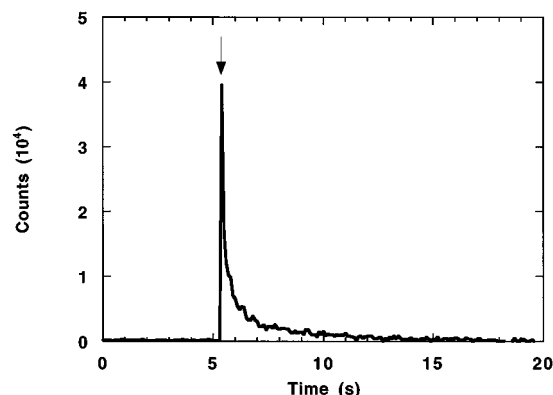


FIG. 2. Laser stimulated thermoluminescence signal obtained from the ZnS:Cu,Nd glass TLD fiber exposed to 4.56 Gy ^{60}Co γ -ray dose and read out using 250 mW cw power at 807 nm.

the rise of the diode laser power and does not at all resemble a traditional glow curve. The explanation of this unique behavior lies in the microscopic nature of the localized laser heating and has been described in detail previously.⁶ Briefly, following optical excitation, the Nd^{3+} ions relax via a combination of radiative and nonradiative pathways. It is expected that Nd^{3+} ions in a Vycor glass matrix will experience efficient nonradiative relaxation due to the high OH concentrations in the glass.⁷ Nonradiative relaxation processes transfer energy to the glass surrounding the absorbing ion, in the form of low frequency vibrational modes of the SiO_2 matrix, resulting in very rapid, localized temperature increases of several hundreds of degrees. For filled traps located very close to the absorbing centers, the thermal barrier to recombination is overcome almost instantaneously. As heat diffuses radially away from the Nd^{3+} ions, the thermal energy is distributed over a much larger volume, and the temperature drops off rapidly. The trap depopulation rate depends on the temperature of the glass relative to the depth of the trap. For traps located further from the absorbing centers, the depopulation rate decreases. The laser reading process used in the present experiments efficiently depopulates the traps as evidenced by the fact that the signal completely returns to the baseline within 10–15 seconds for all exposures studied.

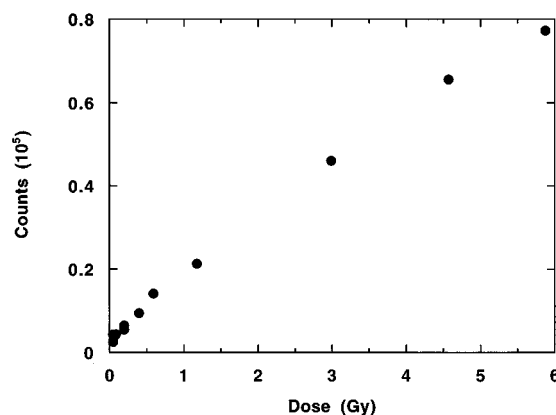


FIG. 3. ^{60}Co γ -ray dose response of the laser-heated ZnS:Cu,Nd glass remote dosimeter. The integrated signal is plotted vs dose.

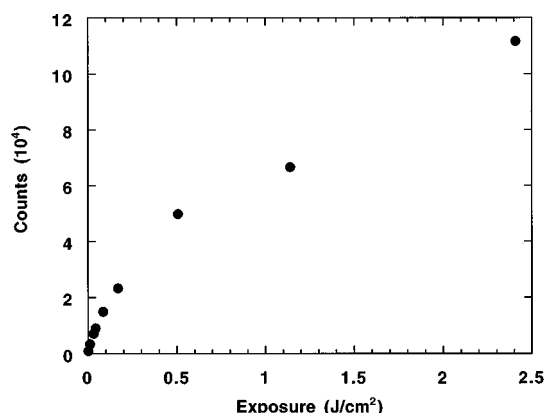


FIG. 4. UV dose response of the laser-heated ZnS:Cu,Nd glass remote dosimeter. The integrated signal is plotted vs dose.

The ^{60}Co γ -ray dose dependence of the fiber dosimeter is shown in Fig. 3. The plot displays the integrated signal counts as a function of the dose, although plots of the peak signal values yield identical dose response curves. The dose dependence is linear for the dose range studied, which includes the dose range of interest for medical diagnostic and radiotherapy applications. The optical transparency of the glass permits increasing the sensitivity and dynamic range of the dosimeter simply by increasing the diameter of the fiber sensor. Medical fiber catheterization techniques can utilize fiber diameters of 1 mm or more. A 3 cm long, 1 mm diam fiber, using the ZnS:Cu,Nd TL glass is expected to be ~ 25 times more sensitive than the 200 μm diam fiber used in this study.

In addition to the γ -ray remote sensing described above, we have also measured the sensitivity of the fiber-optic-coupled remote sensor to ultraviolet (UV) radiation. The ultraviolet sensitivity of a related TL glass has been previously measured and found to be quite good for wavelengths less than 300 nm.⁸ A low pressure mercury arc lamp (UVP Inc., Pen-Ray) was used to determine the UV dose response. The distribution of energy among the wavelengths, as specified by the manufacturer, was 90% at 253.7 nm, 4% at 185 nm, and 6% in the remaining lines. A laser-heated TL glass fiber sensor was placed 7.5 cm from the lamp and exposed to an irradiance of 704 $\mu\text{W}/\text{cm}^2$ for times ranging from 1 to 1600 s. A plot of the UV dose response is shown in Fig. 4. The dose response is linear for exposure times of less than 120 s or 80 mJ/cm^2 . (The UV and γ -ray data were obtained using similar fiber sensor materials, but the diode laser, collection

optics, and detection electronics were different. As such, quantitative comparisons between the two sets of data are not possible.) A variety of manufacturing processes utilize UV sources, such as the curing of polymer coatings on paper products and deep UV photolithographic production of integrated circuits. The fiber-optic-coupled, remote radiation sensor that we have described offers an inexpensive approach for monitoring radiation levels and provides a feedback method for manufacturing quality control.

In summary, we have developed an all optical, fiber-optic-coupled radiation dosimeter that has excellent performance characteristics for remote radiation sensing applications. The enabling feature of the dosimeter is an optically transparent, semiconductor-doped TL glass that exhibits efficient localized laser heating. The sensor utilizes relatively low cost components such as a diode laser array with non-critical frequency stability, a commercial multimode optical fiber, simple optical elements, and direct photodetection. Dose information is correlated with either the signal amplitude or the total integrated count, and minimal signal processing is required. Optical annealing of the dosimeter prepares the sensor for repeat use. The monitoring of radiation levels in ground water supplies around nuclear waste sites could utilize sensor arrays consisting of hundreds of fiber-coupled dosimeters, all interrogated from a central control center, using a single diode laser and photodetector. The fiber-optic dosimeter should also be useful as a process control sensor for industrial applications that utilize radiation sources (ultraviolet to γ ray) such as photolithography and radiation curing of polymers. The small size and the high sensitivity of this fiber-optic radiation dosimeter should be ideal for medical applications such as near-real time, *in vivo*, dose monitoring in patients undergoing radiation therapy.

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